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# Novel Diffusivity Measurement Technique

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# NOVEL DIFFUSIVITY MEASUREMENT TECHNIQUE

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## SUMMARY

A common-path interferometer (CPI) system was developed to measure the diffusivity of liquid pairs. The CPI is an optical technique that can be used to measure changes in the gradient of the refraction index of transparent materials. This system uses a shearing interferometer that shares the same optical path from a laser light source to the final imaging plane. Hence, the molecular diffusion coefficient of liquids can be determined using the physical relations between changes in the optical path length and the liquid phase properties. The data obtained with this interferometer were compared with similar results from other techniques and demonstrated that the instrument is superior in measuring the diffusivity of miscible liquids while keeping the system very compact and robust. CPI can also be used for studies in interface dynamics and other diffusion-dominated-process applications.

## INTRODUCTION

Optical diagnostic techniques have become an integral part of many measurement applications in industrial and research laboratories. In recent years, the point diffraction interferometer (PDI) and its phase-shifted version have been used to measure optical wave fronts for lens testing and combustion and fluid flow diagnostics (refs. 1 to 3). The PDI is considered robust (ref. 4) because it has a common-path design. Another technique, the schlieren, has been used for many years for flow visualization and gives an overall qualitative picture of the variations in the index of refraction within a transparent test section. However, there are advantages in using the shearing interferometer, which has the same capability as the schlieren head. The CPI allows one to easily align the focused light on the interferometer head, and it can be used instead of a schlieren head and a PDI to make quantitative measurements in liquids that have large variations in the index of refraction, as is often the case in interface dynamics studies. This CPI capability is due to its basically being a Wollaston prism in combination with a polarizer and an analyzer; thus, to measure large variations in the index of refraction, one need only to use different Wollaston prisms for each particular experimental condition.

The dynamics of miscible interfaces is an active area of research that has been identified to benefit from experimentation in reduced gravity. Recognized as an important transport phenomenon is the study of the flow patterns by the interface when one liquid is slowly displaced by another such that diffusion plays an important role in the dynamics. It has been suggested that nontraditional stresses in the fluids, resulting from the steep variation in the concentration of the miscible fluids in the mixing zone, might be important in the dynamics. These effects are overwhelmed by the buoyancy-caused flow under terrestrial conditions. The diffusion coefficient (diffusivity) of the miscible fluids is a property that is important to the proper design of experiments. In particular, it determines the desirable range for the speeds of the displacing fluids so that the effect of diffusion is not overshadowed by convective transport of the mass. Petitjeans and Maxworthy (ref. 5) used a variation of Wiener's technique, described in Sommerfeld's textbook on optics (ref. 6), to measure the diffusion coefficient. A narrow laser beam was passed through a quartz cell in which two liquids were diffusing into each other across a horizontal miscible interface. As the beam traversed the test cell, it was bent vertically when it traversed a vertical gradient of concentration. A light detector was then used to measure the deflection of the exiting beam as a function of vertical position. The deflection angle is related to the vertical gradient of the index of refraction and hence to the concentration gradient within the test cell. The average diffusion coefficient was obtained from an analysis of the measured concentration gradient profile. The analysis assumed that the diffusion process is one dimensional and is characterized by a constant value of the diffusion coefficient. The technique used by Petitjeans and Maxworthy (ref. 5) to measure the concentration gradient is a point measurement that requires the light beam and the detector to be traversed through the region of interest. Therefore, instantaneous measurement of the concentration gradient cannot be obtained everywhere near the interface. This limitation can be overcome by using a CPI technique that employs Wollaston prisms to obtain the



distribution of the index of refraction in the mixing region. The specific technique incorporates a Wollaston prism operating in finite fringe mode (ref. 7) to visualize the concentration gradient in the mixing zone in real time. This measurement requires a novel approach in setting the prism at a particular angle with respect to the mixing interface orientation. The diffusivity is then obtained from the measured concentration profiles. The author used the finite mode of fringe arrangement of the CPI (Petitjeans and Maxworthy's technique, ref. 5) and reports herein the measured diffusivity of three combinations of pure silicone oils.

## DESCRIPTION OF CONCEPT AND MEASUREMENT APPARATUS

### Interferometric Model Concept

The optical setup of many shearing interferometers, including the subject CPI, is similar to that of a schlieren system; therefore, the term "schlieren interferometer" has become popular. Among a great number of interferometer systems, an arrangement with a Wollaston prism as the shearing element is most frequently used (ref. 7) in experimental fluid mechanics, gas dynamics, and aerodynamics. This discussion will concern the customized CPI system used for the diffusion coefficient measurements reported herein. In contrast to the schlieren technique, the signal obtained with a CPI results from the change in the optical phase of light waves passing through the transparent object, not by light deflection. Two parallel, laterally sheared light rays passing through the test section at coordinates  $(y + d/2)$  and  $(y - d/2)$  are considered (ref. 7), where  $d$  is a small distance between the two rays. The shearing element positioned in or near the focal point of the schlieren head ensures that the two rays coincide after they have passed through this element. Under the assumption that the conditions of optical coherence are fulfilled, the two coinciding light rays can interfere with each other. If the refractive indices  $n$  (or liquid concentration  $c$ ) at coordinates  $(y + d/2)$  and  $(y - d/2)$  in the test section are not equal, two light rays (or waves) exhibit a difference in their optical paths  $\Delta l(x, y)$  as given by

$$\Delta l(x, y) = \int_{z_1}^{z_2} n(x, y + \frac{d}{2}, z) dz - \int_{z_1}^{z_2} n(x, y - \frac{d}{2}, z) dz \quad (1)$$

Two assumptions have been made for deriving this equation: first, the coordinates where the rays enter and leave the test section  $z_1$  and  $z_2$  are equal for the two rays; that is, the test section is bounded by plane walls. Second and more important, the propagation of light in the test section is straight and not deflected. This assumption is reasonable for most gas flows and for very low to moderately sheared liquids. One should be aware that this assumption might be violated in some liquid flows and gases with very strong density gradients (e.g., in plasma flows). Exceptions from the validity of this assumption, known as "strong refraction," will not be of concern here. Strong refraction is much more complicated because neither the refractive index (the concentration or density) nor the path of the light in the test section is known.

In the present CPI system, the shear or the separation of the two considered light rays  $d$  is assumed to be very small in comparison with the dimensions of the field of view. The right-hand side of equation (1) can be developed into a Taylor series around  $n(x, y, z)$ . Ignoring the higher order terms and taking into account only the linear term (because  $d$  is small) yields

$$\Delta l(x, y) = d \int_{z_1}^{z_2} \frac{\partial}{\partial y} n(x, y, z) dz \quad (2)$$

The quantity  $\Delta l(x, y)$  is measured in the recording plane in the form of bright interference fringes that appear where

$$\frac{1}{\lambda} \Delta l(x, y) = 0, \pm 1, \pm 2, \dots \quad (3)$$



with  $\lambda$  being the light wavelength. The equation for the dark fringes is

$$\frac{1}{\lambda} \Delta l(x, y) = 0, \pm \frac{1}{2}, \pm \frac{3}{2}, \dots \quad (4)$$

Note that the measured signal, the left-hand side of equations (3) and (4), is two dimensional whereas the quantity to be determined  $n(x, y, z)$  is three dimensional in the general case. A direct measurement of  $n$  is possible for a plane refractive index field  $n(x, y)$ .

The dependence of the measured signal on the gradient of the refractive index, as in the techniques that rely on

the measurement of the deflection angle  $\varepsilon = \frac{n}{n_a} \int_{z_1}^{z_2} \frac{1}{n} \frac{\partial n}{\partial y} dz$  (described in ref. 6), can be seen in equation (2), where  $n_a$  is the refractive index of air. The measurement of  $\partial n / \partial x$  is also possible by rotating the optical system  $90^\circ$  around the optical axis ( $z$ -axis in this case). From equation (2) it follows that an interferogram with  $\partial n / \partial y$  will appear uniformly illuminated; all light waves arrive in the recording plane with the same optical phase. This case is called the "infinite fringe mode" alignment of the interferometer. It can be aligned in a different way so that a set of equidistant parallel interference fringes appear for the object with  $\partial n / \partial y = \text{a constant}$ .

For the study reported herein, the interferometer was set in the finite fringe mode, and the prism was rotated around the optical axis  $z$  to create a small angle between the fringes and the diffusing interface under investigation, making it easy to illustrate the concept that the gradient of the index of refraction is directly proportional to the concentration gradient in the fluids (ref. 8). The final relations are given in the following equations (ref. 8).

For  $n = n(y)$ ,

$$\frac{\partial n}{\partial y} = K \frac{\partial c}{\partial y} \quad (5)$$

And similarly for  $n = n(x)$ ,

$$\frac{\partial n}{\partial x} = Q \frac{\partial c}{\partial x} \quad (6)$$

where parameters  $K$  and  $Q$  are constant values that are proportional to the wavelength and inversely proportional to the object length  $L$ , the separation distance  $d$ , and the angle between the interface of miscible fluids under measurement and the fringe direction. Equations (5) and (6) do show that the concentration gradient is proportional to the refractive index gradient. By using this concept and assuming that the diffusion process is one dimensional and is characterized by a constant value of the diffusivity as described in the section Diffusivity Calculation, the diffusivity for two miscible liquids in contact was obtained.

#### Test Apparatus and Procedure

Upon using the concept expressed in equations (5) and (6) from reference 8, the CPI measures the gradient of the index of refraction in a certain direction within a test cell. A Wollaston prism was used to image the sheared light beam by the concentration gradient near a miscible interface. A collimated, polarized beam of light from a low-power laser is passed through a transparent test cell. The light is then focused on the Wollaston prism, which splits the light into two beams that are slightly displaced from each other. When the beams are recombined, they produce interference fringes that indicate gradients of refraction in the test medium. The interferometer is arranged in its finite fringe mode where equidistant, parallel interference fringes appear in a uniform index-of-refraction field in the test cell. This is the case when only one of the fluids is present in the test cell. When a second liquid is introduced and diffusion occurs, the deviation or shift of a fringe from its undisturbed location is a measure of the index-of-



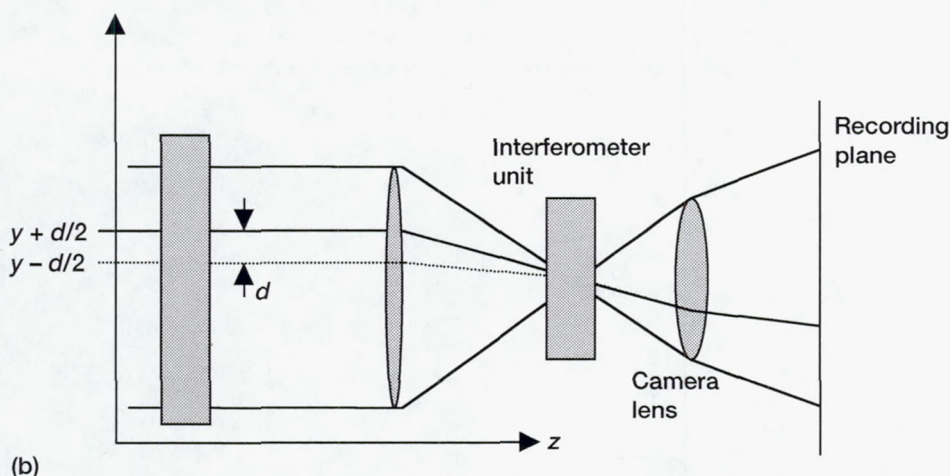
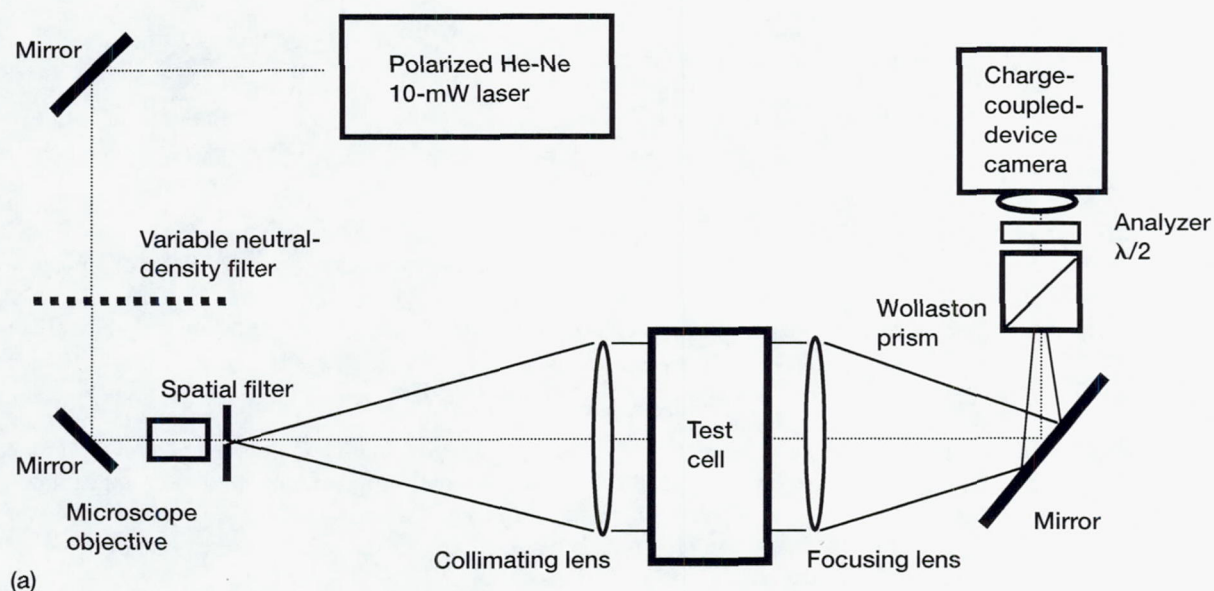


Figure 1.—Test apparatus. (a) Common-path interferometer (CPI). (b) Expanded shearing unit.

refraction gradient within the test cell. It has been assumed that the index of refraction within the test cell is only a function of the vertical coordinate. The light beam is passed through the test cell horizontally, and it has been further assumed that each ray of light traverses a path of constant refractive index without refraction. Figure 1 shows a schematic of the CPI and the interferometer unit. The test cell used for the diffusivity measurement of a pair of miscible liquids is a quartz container 10 by 10 mm in cross section and 45 mm in height (fig. 2). A charge-coupled-device (CCD) camera captured the interferograms of the CPI and a time-lapse super video recorder (S-VHS) stored them on a tape for later analysis. Figure 3 shows a typical fringe image near the diffusing interface after the fluids came in contact. Figure 4 illustrates the trace of a typical fringe near the interface under investigation and also shows the data points that are easily identified and used for calculating the diffusivity of the liquids under measurement. These points are selected because they simplify the diffusivity calculations when an average of the diffusivity over some length of time is needed (explained in the next section). The fluids used in this measurement are silicone oils of various viscosities (table I).

#### Diffusivity Calculation

When two miscible liquids are put in contact, the two fluids are assumed to be pure and therefore only binary diffusion occurs. Let  $\rho_1(x,t)$  and  $\rho_2(x,t)$  represent the mass concentration of the two fluids at a vertical location  $x$  and



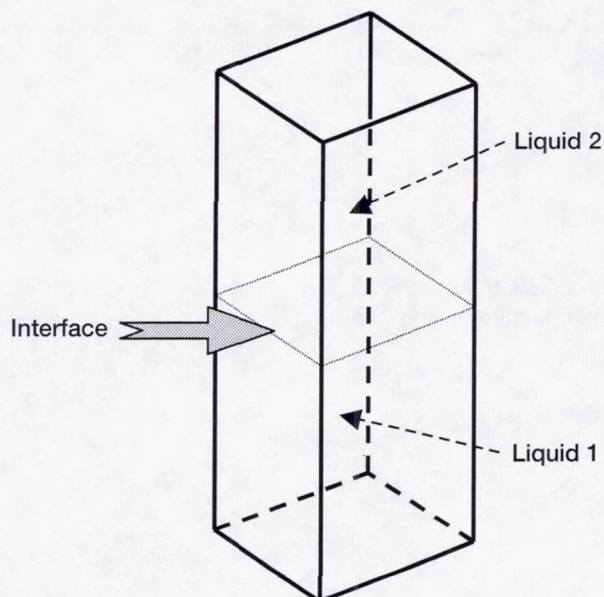


Figure 2.—Quartz test container (10 by 10 by 45 mm).

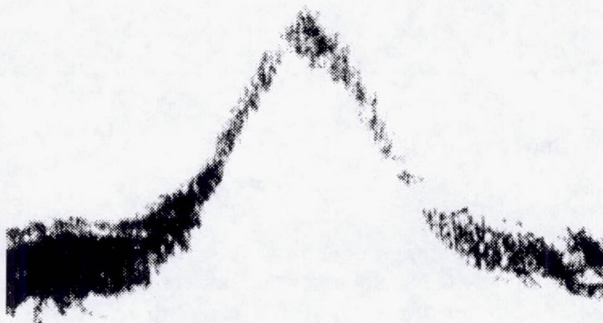


Figure 3.—Typical fringe near interface 30 min after liquids came in contact. (Note: The rest of fringes near the interface are not shown because their spacing is larger than the shift peak at this moment.)

at a given time  $t$ . Far away from the interface,  $\rho_{1\infty}$  and  $\rho_{2\infty}$  are the densities of the two fluids. It is convenient to define the dimensionless molar concentration of liquid 1 as  $c = \frac{\rho_1(x)}{M_1\rho_{1\infty}}$ , where  $M_1$  is the molecular weight of liquid 1. A similar dimensionless concentration for liquid 2 may be defined. For stationary liquids, the governing equation for diffusion is (ref. 9)

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (7)$$

Strictly speaking, this equation is valid only when the binary diffusion coefficient  $D$  and the local density in the liquid are constant. The coefficient of diffusion from our experiment will be extracted assuming that equation (7) is valid. The boundary conditions are

$$c \rightarrow 1 \text{ as } x \rightarrow \infty \quad \text{and} \quad c \rightarrow 0 \text{ as } x \rightarrow -\infty \quad (8)$$

The solution for the concentration gradient is

$$\frac{\partial c}{\partial x} = \frac{1}{2\sqrt{\pi Dt}} e^{-\left(x^2/4Dt\right)} \quad (9)$$

The dimensionless concentration gradient profile for liquid 2 is identical to that for liquid 1. Assuming that the refractive index is linearly proportional to the concentration of the two fluids, the gradient of the refractive index has a similar profile.

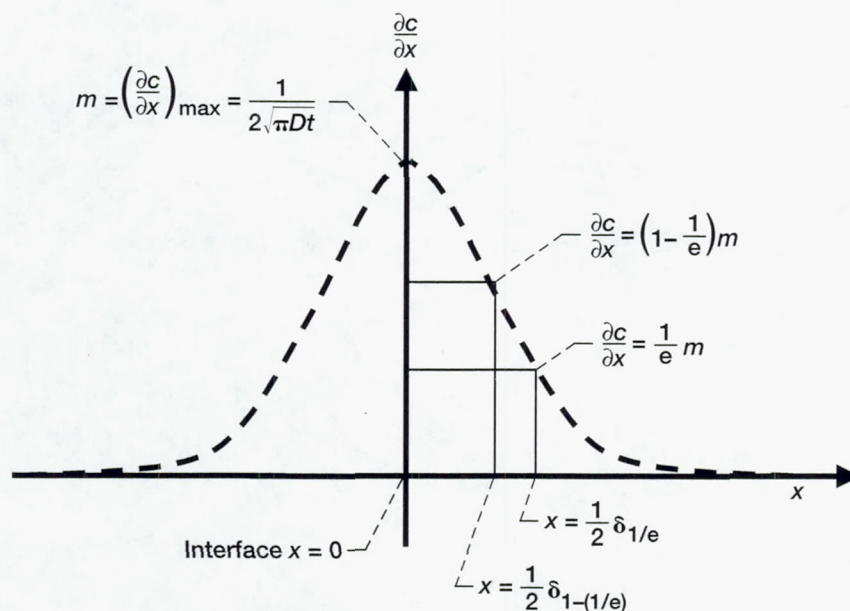


Figure 4.—Typical trace of fringe near interface after liquids came in contact. Data points for diffusivity  $D$  calculation are also shown on traced shifted fringe. (Note: All dimensions are normalized by the value of concentration gradient  $m$ .)

TABLE I.—COMPARISON OF AVERAGED DIFFUSIVITIES

Silicone oil liquid pairs, cSt	Averaged diffusivities, $\text{cm}^2/\text{s}$			Technique
	$D_{1-(1/e)}$	$D_{1/e}$	$D_{\text{Wiener}}$	
1000/100	$(4.36 \pm 0.30) \times 10^{-8}$	$(4.79 \pm 0.22) \times 10^{-8}$	$(1.69 \pm 0.06) \times 10^{-8}$	Wiener (ref. 10)
	$(5.11 \pm 0.73) \times 10^{-8}$	$(5.41 \pm 0.75) \times 10^{-8}$	-----	Common-path interferometer
1000/10	$(2.61 \pm 0.10) \times 10^{-7}$	$(2.47 \pm 0.10) \times 10^{-7}$	$(3.14 \pm 0.15) \times 10^{-7}$	Wiener (ref. 10)
	$(2.83 \pm 0.72) \times 10^{-7}$	$(2.34 \pm 0.68) \times 10^{-7}$	-----	Common-path interferometer
1000/1	$(4.17 \pm 0.23) \times 10^{-6}$	$(2.48 \pm 0.21) \times 10^{-6}$	$(1.93 \pm 0.08) \times 10^{-6}$	Wiener (ref. 10)
	$(2.65 \pm 0.32) \times 10^{-6}$	$(1.93 \pm 0.29) \times 10^{-6}$	-----	Common-path interferometer



The maximum value of the concentration gradient is at  $x = 0$  and is given by

$$m = \left( \frac{\partial c}{\partial x} \right)_{\max} = \frac{1}{2\sqrt{\pi Dt}} \quad (10)$$

For positive  $x$ , let  $x = \frac{1}{2} \delta_{1-(1/e)}$  at the location where  $\frac{\partial c}{\partial x} = \left(1 - \frac{1}{e}\right)m$ . The solution given for the concentration gradient can then be used to obtain the diffusivity:

$$D_{1-(1/e)} = \frac{\delta_{1-(1/e)}^2}{\left[ -16 \ln \left( 1 - \frac{1}{e} \right) \right] t} = \frac{\delta_{1-(1/e)}^2}{7.334t} \quad (11)$$

Similarly, let  $x = \frac{1}{2} \delta_{1/e}$  at the location where  $\frac{\partial c}{\partial x} = \frac{1}{e}m$ . Then  $D_{1/e}$  is obtained to be

$$D_{1/e} = \frac{\delta_{1/e}^2}{16t} \quad (12)$$

The values of  $\delta_{1-(1/e)}$  and  $\delta_{1/e}$  have been obtained from the measured refractive index gradient profiles. The time  $t$  after the fluids came in contact and each value of  $\delta_{1-(1/e)}$  or  $\delta_{1/e}$  extracted from the recorded fringes (fig. 4 shows a typical fringe profile with the data points of interest on it) can be used to calculate the diffusivity from equations (11) and (12), respectively. For an average diffusivity, the values of  $\delta_{1-(1/e)}$  and  $\delta_{1/e}$  for various values of  $t$  were used and  $\delta^2$  was plotted versus  $t$ . The values for diffusivity, with its 95-percent confidence interval, were obtained by using linear regression and finding the best fitting straight line to such plots.

## RESULTS

Table I shows the measured diffusivity (averaged over time) for several silicone oil liquid pairs. For comparison, it also contains a set of data similar to that obtained with other techniques (ref. 10). The diffusivity was obtained with the assumption that the simple theory of binary diffusion holds. In all pairs of liquids tested, from the regression analysis, a 95-percent confidence interval for the measured diffusivity is reported herein. The diffusivity measured by the original Wiener's technique reported in reference 10 appears to be the least accurate, perhaps because the refractive index must be known more precisely for the wavelength of the light used. In the case of diffusion across the interface of one silicone oil pair (1 and 1000 centistokes), the difference in diffusivity obtained by the various techniques is large (36 percent because of the nongaussian distribution in the concentration gradient across the interface). This pair has the largest density difference ( $\Delta\rho/\rho = 0.19$ ) among the various techniques compared herein. One possible explanation for the gap between data sets is that the simple model of diffusion that was assumed may not hold for this pair.

The reliability of the CPI has made it attractive for application in diffusion measurements near moving interfaces of miscible fluids in selected space experiments (ref. 11).



## CONCLUSIONS

A common-path interferometer (CPI) was developed and used in its finite fringe mode. A novel approach was taken to provide a fringe profile that resembled the solution to a simple model of a diffusion equation. The approach involved setting a small angle between the fringe orientation and the interface of the two miscible liquids. Then a mathematical model of the optics for this situation was given. From the analysis of the fringe profile obtained in this manner, the diffusion coefficients (diffusivity) of pairs of miscible fluids were calculated.

The technique used herein was compared with Wiener's original technique, which requires the values for the refractive indices of the two miscible fluids. His appears to be the least accurate because the refractive indices must be known more precisely for the wavelength of light used. This information is not required for the CPI technique. The diffusion coefficients, measured by interferometry and a modified Wiener's technique that analyzes the measured concentration gradient profiles, are in good agreement. The exception is the case of diffusion across the interface of a silicone oil pair of 1 and 1000 centistokes, for which the difference is as much as 36 percent. This pair has the largest density difference. An explanation might be that the simple model of diffusion used herein to extract the diffusion coefficient did not strictly hold for this pair. The measured concentration gradient profile is not a gaussian symmetric profile. Also the diffusivity may be dependent on the concentration itself, which requires further analysis of the data to extract such information from similar interferometric signals.

The approach reported herein and the application of the technique to double-diffusive problems will be the subject of future studies. The present CPI technique has been adopted for diffusion measurement near moving interfaces of miscible fluids in a selected space experiment in the near future.

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